Urbanization is among the leading causes of increased air particulate pollution in Southeast Asia. This situation puts Calaca, Batangas – a first-class rural municipality transitioning to a renowned industrial hub – at risk of the harmful effects of air particulate matter (PM). To determine the composition and possible sources of air pollution in Calaca, ambient air particulate samples were collected from December 2018 to March 2019. A total of 52 filter samples, 26 each from the PM$_{10-2.5}$ and PM$_{2.5}$ fractions, were collected and analyzed for PM$_{10}$, PM$_{2.5}$, multiwavelength black carbon (BC), and elemental concentrations. Average PM$_{10}$ and PM$_{2.5}$ mass concentrations were 34.01 ± 12.03 μg/m$^3$ and 10.62 ± 4.86 μg/m$^3$, respectively. These concentrations comply with the annual National Ambient Air Quality Guideline Values (NAAQGV) of the Philippine Clean Air Act of 1999 but exceed the prescribed guideline value of the World Health Organization (WHO). Results from energy-dispersive x-ray fluorescence (EDXRF) spectroscopy, a nuclear analytical technique, showed 14 elements present in the PM$_{2.5}$ fraction – namely S, Si, K, Fe, Ca, Al, Cl, Mn, Na, Ti, Zn, Ni, Sc, and Cu arranged in decreasing average concentrations. Average biomass (BBC) and vehicular black carbon (VBC) concentrations from multiwavelength analysis were 0.7 ± 0.3 μg/m$^3$ and 0.7 ± 0.6 μg/m$^3$, respectively. By reconstructing masses using these data, contributions of different sources of air PM were determined to be salt (0.54%), soil (11.31%), (NH$_4$)$_2$SO$_4$ (61.15%), VBC (6.33%), and BBC (6.28%). Conditional probability function (CPF) determined that sources of PM are likely located at the 30–60° and 180° directions from the sampling site. This study provides baseline data for the chemical composition and source identification of PM in Calaca, Batangas.
Urbanization, along with exponential human and vehicle population growth, is among the leading causes of increased air pollution in Southeast Asia (Kim Oanh et al. 2006). Epidemiological studies and demographic data show that air pollution consistently correlates to an increase in mortality rate and an increase in hospital admissions due to respiratory and cardiovascular diseases (Brunekreef and Holgate 2002). One primary measure of air pollution is airborne PM, and the Philippines has among the highest concentrations of PM in the region (Hopke et al. 2008).

PM is a complex mixture of organic and inorganic substances in the form of solid particles and liquid droplets emitted into the atmosphere through natural (e.g. volcanic eruptions, dust storms, sea spray) or anthropogenic means (e.g. fuel combustion, cooking, emissions from humidifiers) (Taner et al. 2013). PM is usually classified according to particle size. Particles with an aerodynamic diameter of ≤ 10 µm are classified as PM$_{10}$, while those with an aerodynamic diameter of ≤ 2.5 µm are classified as PM$_{2.5}$ (Donaldson et al. 2000, Peng et al. 2008). PM$_{10}$ can be further subdivided into fractions. Particles with an aerodynamic diameter ranging from 2.5–10 µm comprise the coarse or inhalable fraction (PM$_{10-2.5}$), while those with < 2.5 µm diameter comprise the fine or respirable fraction (Pabroa et al. 2011). PM can induce harmful health effects to the human body when its particle size is small enough to be inhaled and penetrate our respiratory tract (Guo et al. 2018). Long-term exposure to PM may lead to different illnesses related to cardiovascular, inflammatory, and respiratory issues. Such respiratory diseases include chronic obstructive pulmonary disease, asthma, different types of lung infection and inflammation, and even lung cancer (He et al. 2017). Aside from the particle size, the chemical composition of PM also influences its adverse effects on human health. The presence of heavy metals in PM is also widely monitored due to its extreme health risks, even in minute concentrations (Duruibe et al. 2007). Generally, these heavy metals are known to intercalate with DNA and displace original metals from their natural protein binding sites, resulting in the oxidative deterioration of cells and other biological macromolecules (Jaishankar et al. 2014).

Carbonaceous compounds, sometimes generally referred to as black carbon, is another component of PM. Its ability to absorb light makes it a potent climate forcing agent that contributes to the heating of Earth’s atmosphere, especially the lower and middle troposphere (Singh et al. 2018). It is also known to induce potential toxicological effects on the human body, especially to the cardiovascular and respiratory systems (Kecorius et al. 2017). BC is primarily produced by the incomplete combustion of carbon-containing fuels such as coal, biomass fuel, and diesel (Gertler et al. 2016). Processes related to the burning of these fuels include vehicle use, electric power production by power plants, and other industrial productions. These processes are significantly present in countries undergoing urbanization (Paredes-Miranda et al. 2013).

As far as we know, there is no available information regarding the existing disposal and emission means of the industries around Calaca. Moreover, most industries, especially small-scale ones, do not have proper waste treatment facilities and lack pollution control equipment (Blackman et al. 2006). This situation puts the population of Calaca and its nearby municipalities at risk of the harmful effects of PM. Therefore, there is a need to obtain baseline information about the levels, composition, and possible sources of air PM in Calaca, Batangas.

The primary objective of this study is to provide preliminary data about the composition and possible sources of air PM in Calaca, Batangas. EDXRF spectroscopy, a nuclear analytical technique, was the primary tool used for multi-element analysis of the acquired particulate samples. Moreover, gravimetric and multiwavelength analyses were employed to determine PM and BC concentrations, respectively. Possible geographical sources of PM were also determined using CPF to provide inferences about the possible sources of the pollution in the area. Integrating all these data, mass reconstruction was done to determine possible sources and their respective contributions to the total airborne PM levels in Calaca, Batangas. The baseline information generated in this study will help the local government and concerned agencies to make and improve environmental policies and regulations for the protection of the population from the harmful effects of air pollution.

**MATERIALS AND METHODS**

Air particulate samples were collected in a residential area in Brgy. Camastilisan, Calaca, Batangas, Philippines (13°55'48.6" N 120°48'23.4" E; Figure 1). The chosen sampling site is approximately 2 km from the SEM-Calaca Coal-Powered Plant, 2 km from the industrial complex of Calaca Seaport, Inc., 0.1 km from the national highway, and 0.8 km from the shoreline.

A Gent dichotomous sampler was placed 3.5 m above ground level, which collected coarse (PM$_{10-2.5}$) and fine (PM$_{2.5}$) PM samples on Nucleopore polycarbonate filters (Hopke et al. 1997). Air sampling was performed twice a week on Wednesdays and Sundays to represent weekday and weekend levels of the parameters measured. A total of 26 samples each for the PM$_{10-2.5}$ and PM$_{2.5}$ fraction were
collected throughout the sampling period of four months (December 2018 to March 2019). Filters were weighed before and after sampling using Mettler MT5 analytical microbalance with 1 ug sensitivity. Before every weighing, filters were equilibrated for at least 24 h at constant temperature and humidity. PM$_{10}$ and PM$_{2.5}$ concentrations were then calculated from the measured net weights for each filter using Equation 1. Data samples that did not have 12.5–15.0 m$^3$ of the total volume of air sampled were omitted because this indicates that sampling error occurred.

$$PM\text{ conc.} = \frac{\text{net weight (ug)}}{\text{volume of air sampled (m}^3\text{)}}$$ (1)

Elemental analysis of the PM$_{2.5}$ samples was performed in the Nuclear Analytical Techniques Application Laboratory of DOST-PNRI, Diliman, Quezon City using Panalytical Epsilon 5 EDXRF spectrometer. The spectra were processed and analyzed using the built-in Qualitative X-Ray Analysis System. Calibration was performed using SRM 2783, a standard reference material (SRM) from the National Institute of Standards and Technology (NIST). SRM 2783 is an air particulate sample (PM$_{2.5}$) deposited on a polycarbonate filter similar to the real samples and is primarily intended for the calibration of methods of analysis for the elemental composition of airborne PM$_{2.5}$ in an urbanized industrial area (Lim et al. 2010, NIST 2011).

Elemental concentrations obtained from EDXRF analysis were used to estimate concentrations of the source types likely to contribute to the measured elements. The reconstructed masses (RCM) of the particular matter samples were calculated by determining the concentration of the major aerosol species (i.e., salt, ammonium sulfate, soil, VBC, and BBC) from the standard defined by Malm et al. (1994).

Furthermore, possible sources of BC in PM$_{2.5}$ samples were determined using multi-wavelength absorption black carbon instrument (MABI), an instrument capable of measuring light absorption in a filter at seven different wavelengths: 405 nm, 465 nm, 525 nm, 639 nm, 870 nm, 940 nm, and 1050 nm. As reported by the Australian Nuclear Science and Technology Organization, VBC and BBC concentrations were calculated from the obtained absorbance values at 639 nm and 405–1050 nm, respectively.

Calculation and statistics of the data from the study were processed using Microsoft Office® Excel 2019 and Statgraphics® Centurion XVIII. Obtained PM concentrations coupled with wind direction values on-site obtained from http://www.timeanddate.com (Time and Date n/d) were utilized to locate possible sources of these species via CPF (Kim and Hopke 2004).

RESULTS AND DISCUSSION

Levels of PM in Calaca, Batangas

A time series plot was created for the twenty-six (26) samples of both PM$_{10}$ and PM$_{2.5}$ fractions, as shown in Figure 2. Both time-series plots include annual and daily acceptable concentration values for both PM fractions set.
Figure 2. Time series plot for (a) PM\(_{10}\) and (b) PM\(_{2.5}\) concentrations (μg/m\(^3\)) in Calaca, Batangas from December 2018 to March 2019.

Figure 3. Upper 50% CPF graphs for (a) PM\(_{10}\) and (b) PM\(_{2.5}\) obtained from the wind flow behavior of Calaca, Batangas.

Figure 4. BC concentrations at (a) 405–1050 nm and (b) 639 nm, and their (c) comparative graph that corresponds to vehicular emission (639 nm) and biomass burning (405–1050 nm) as sources of BC.

by the WHO and the Philippine Clean Air Act of 1999. Samples from Feb 10 and 13 were not included in the graph due to sampling error made by the operator.

CPF
To further determine the possible sources of PM, CPF was used. It is a tool that estimates the probability that a given concentration will exceed a threshold from a given direction (Kim and Hopke 2004). In this study, the upper 50\(^{th}\) percentile – instead of the usual upper 25\(^{th}\) percentile – was used as the threshold criterion due to the small sampling size. CPF indicates possible sources of PM in the 30–60\(^{o}\) direction or northeast of the sampling site, which directly points toward the highway. Another possible source comes from the south or 180\(^{o}\) direction, which points to the sea (Figure 3).

Due to the possible detrimental effects of PM\(_{2.5}\) once it penetrates the inferior airways, PM\(_{2.5}\) samples were further analyzed to determine its BC and elemental concentrations.

Multiwavelength Absorption Analysis of BC
Only three wavelengths were considered for the discussion: 639 nm, 405 nm, and 1050 nm. Values obtained from 1050 nm were subtracted from the values from 405 nm, and the difference represents the BC concentrations contributed by biomass burning (Figure 4a). Values from 639 nm were
directly plotted as it represents the BC concentrations contributed by vehicular emission (Figure 4b) and both graphs were transformed and compared using a time series plot as shown in Figure 4c.

Elemental Composition and Chemical Concentrations of PM

A total of 14 elements were detected in the PM$_{2.5}$ fraction using EDXRF: Na, Al, Si, S, Cl, K, Ca, Sc, Ti, Mn, Fe, Ni, Cu, and Zn. The mean, minimum, and maximum concentrations of the elements along with the BC (VBC and BBC) concentrations from MABI are shown in Table A4 (see Appendix III). A box-and-whiskers plot was also used to present the distribution of the elements detected in the PM$_{2.5}$ fraction, as shown in Figure 5. Sulfur was found to be most abundant, having an average concentration of 1.573 μg/m$^3$ and concentrations up to 3.074 μg/m$^3$ (see Appendix III). Elements typical of soil-related PM such as Al, Si, Ca, Ti, and Fe were also present in small concentrations. K and Mn were also found in the samples, which are elements associated with smoke and traffic emissions (Cohen 2010). The presence of heavy metals was not observed in the PM$_{2.5}$ fraction.

Source Apportionment

Correlation analysis was performed to determine and verify common source types of the detected elements in the PM$_{2.5}$ fraction. A correlation matrix was used to determine correlated elements, as shown in Table 1. A strong positive correlation was established between the five soil components: Al, Si, Ca, Ti, and Fe. The most abundant element in the fraction, S, was correlated well

![Figure 5. Box-and-whiskers plot of elements detected in the PM$_{2.5}$ fraction.](image)

**Table 1.** Correlation matrix of the elemental concentrations in the PM2.5 fraction (highlighted values represent $r$-values $> 0.60$ and $r$-values $> 0.50$).

<table>
<thead>
<tr>
<th></th>
<th>Na</th>
<th>Al</th>
<th>Si</th>
<th>S</th>
<th>Cl</th>
<th>K</th>
<th>Ca</th>
<th>Sc</th>
<th>Ti</th>
<th>Mn</th>
<th>Fe</th>
<th>Ni</th>
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<td>Na</td>
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<td></td>
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<tr>
<td>Al</td>
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<td></td>
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<td></td>
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<tr>
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<td>0.58</td>
<td>0.38</td>
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<tr>
<td>K</td>
<td>-0.11</td>
<td>0.79</td>
<td>0.94</td>
<td>0.73</td>
<td>0.16</td>
<td>0.58</td>
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<td>-0.37</td>
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<td></td>
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<tr>
<td>Sc</td>
<td>0.30</td>
<td>0.88</td>
<td>0.88</td>
<td>0.65</td>
<td>0.31</td>
<td>0.57</td>
<td>0.87</td>
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<td></td>
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<tr>
<td>Ti</td>
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<td>0.30</td>
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<td>0.21</td>
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<td></td>
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<tr>
<td>Fe</td>
<td>-0.11</td>
<td>0.87</td>
<td>0.97</td>
<td>0.68</td>
<td>0.25</td>
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<td>0.91</td>
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<td></td>
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<td>-0.11</td>
<td>0.06</td>
<td>0.02</td>
<td>-0.06</td>
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<td>-0.04</td>
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<tr>
<td>Ni</td>
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<td>0.02</td>
<td>-0.05</td>
<td>0.03</td>
<td>0.26</td>
<td>0.64</td>
<td>0.01</td>
<td>0.07</td>
<td>-0.00</td>
<td>-0.15</td>
<td>-0.06</td>
<td>0.09</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cu</td>
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<td>0.50</td>
<td>0.53</td>
<td>0.74</td>
<td>0.14</td>
<td>0.42</td>
<td>0.38</td>
<td>-0.31</td>
<td>0.66</td>
<td>0.33</td>
<td>0.63</td>
<td>0.12</td>
<td>-0.07</td>
<td></td>
</tr>
<tr>
<td>Zn</td>
<td>0.50</td>
<td>0.53</td>
<td>0.74</td>
<td>0.14</td>
<td>0.42</td>
<td>0.38</td>
<td>-0.31</td>
<td>0.66</td>
<td>0.33</td>
<td>0.63</td>
<td>0.12</td>
<td>-0.07</td>
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</tr>
</tbody>
</table>
with the terrestrial elements (i.e., Al, Si, Ca, Ti, and Fe), and an anthropogenic element (i.e., Zn). Correlations between S-Mn and S-K were also observed, typical of industrial emissions.

Malm et al. (1994) devised a standard way to estimate the significant components (e.g., salt, ammonium sulfate, soil, smoke) of aerosol mass through pseudo-elemental calculations or the use of the obtained elemental concentrations to determine the estimated contributions of the components mentioned above to the particulate fraction. Results from this method showed that the largest contributor to the total mass of the PM$_{2.5}$ fraction was (NH$_4$)$_2$SO$_4$, with an estimated contribution of 61.15%, while salt had the lowest contribution of 0.54%. The observed contributions of VBC and BBC in the PM$_{2.5}$ fraction were 6.33% and 6.28%, respectively. The soil component of the PM$_{2.5}$ fraction was determined to contribute a total of 11.31% of the total PM$_{2.5}$ mass. Upon calculation of reconstructed mass, it was determined that source contributions account for the 85.61% of the total PM$_{2.5}$ (Figure 6). The unapportioned component (labeled as “others”), which represents the difference between the average PM$_{2.5}$ concentration and the sum of the average source contributions from the mass reconstruction, accounts for 14.39% of total PM$_{2.5}$.

A time series plot for the apportioned PM$_{2.5}$ sources of the twenty-six (26) PM$_{2.5}$ samples was also constructed along with the calculated individual PM$_{2.5}$ concentrations of each sample.

Identified PM$_{2.5}$ sources from the mass reconstruction showed variations in the source emission strengths, as shown in Figure 7. For example, the unapportioned mass, labeled as “others,” has some significant peaks, specifically on 30 Jan 2019. Overall, sulfate contributions comprise the largest fraction of most of the PM$_{2.5}$ samples, with concentrations up to 12.68 µg/m$^3$.

**DISCUSSION**

**Baseline Measurement of PM$_{10}$ and PM$_{2.5}$**

Results showed that PM$_{10}$ concentrations in Calaca, Batangas ranged from 4.64 to 61.92 µg/m$^3$, with an average of 34.01 µg/m$^3$ (Figure 2a). This average PM$_{10}$ concentration was compared to the annual PM$_{10}$ concentration guideline values set by the Philippine Clean
Air Act and WHO. Average PM$_{10}$ concentration in Calaca complied with the NAAQGV of the Philippine Clean Air Act of 1999 (60 μg/m$^3$) but exceeded the guideline values set by the WHO (20 μg/m$^3$) by 70%. This finding means that PM$_{10}$ levels in Calaca follow the local standard set by the Philippines, which is arguably a more comparable standard due to geographic parameters (e.g., wind flow) that could differ when measured internally compared to that of local measurements. Individual PM$_{10}$ concentrations were considered safe and below the limits set by the WHO and NAAQGV except for PM$_{10}$ concentrations on 19 Dec 2018 and 10 Mar 2019, which have concentrations of 61.92 and 50.75 μg/m$^3$, respectively. These daily concentrations are both higher than the WHO 24-h standard of 50 μg/m$^3$. It was shown that the lowest PM$_{10}$ concentration was obtained on 30 Dec 2018. On that particular date, observations reported that it rained in the sampling site for almost 24 h. Weather data retrieved from an online weather forecasting outlet recorded rain occurring on 28–31 Dec 2018, with 17–53 mm of precipitation (Accuweather n/d). Studies show that rain could significantly decrease the amount of PM in the atmosphere due to the washing effect induced by precipitation (Ouyang et al. 2015).

For PM$_{2.5}$, results showed a concentration range of 2.63–22.27 μg/m$^3$, with an average of 10.62 μg/m$^3$ (Figure 2b). The obtained average PM$_{2.5}$ concentration slightly exceeded the guideline values set by WHO (10 μg/m$^3$) while still under the NAAQGV of the Philippine Clean Air Act of 1999 (25 μg/m$^3$). Individually, all PM$_{2.5}$ concentrations complied with the NAAQGV and the values set by WHO with some peaks on 30 Jan 2019 and 17 Mar 2019. Temporal % analysis of the possible PM$_{2.5}$ sources (Figure 7) shows that the peak on 30 Jan 2019 is accompanied by a significant increase in the unapportioned component of PM$_{2.5}$ but a decrease in the sulfate component. This finding indicates that the observed peak on 30 Jan 2019 is not influenced by the sources of sulfate such as fuel combustion from vehicular emissions, but is driven by the unapportioned mass – possibly organic components – that could not be identified and measured by the source apportionment model. The peak on 17 Mar 2019, on the other hand, is accompanied by an increase in the soil component of the reconstructed mass, along with the unapportioned component of PM$_{2.5}$. This increase may have been caused by activities that bring soil dust from the ground into the atmosphere, such as wind and movement of anthropogenic origins. Similar to the PM$_{10}$ fraction, it was also shown that the PM$_{2.5}$ fraction had its lowest concentration on 30 Dec 2018, possibly due to the washout effect by the rain that occurred on that sampling date.

Weekday and weekend PM concentrations, for both PM$_{10}$ and PM$_{2.5}$, were analyzed and compared using T-Test with a confidence level of 95%. This was done to determine possible sources of PM that contribute to the area. However, no significant differences were found out when monthly weekends and weekdays were compared (Appendix II).

**CPF**

In order to visualize possible sources of PM in the area, the result of the CPF was overlain on a map of the sampling site (Figure 8).

CPF points towards the 30–60° direction, in which a national highway is located. This finding suggests that for the duration of our sampling, PM mainly came from...
vehicular activity coming from the national highway. This explanation is supported by the presence of VBC and high amounts of sulfur, possibly also coming from vehicular emissions. Another possible source of PM from this direction is soil dust caused by the movement of residents accessing the highway, and because the ground in this area is uncremented.

CPF indicates that PM also mostly likely comes from the 180° direction, which points to the sea. Possible PM sources from this direction are ship emissions, which are characteristic of areas where ship traffic is present (Contini et al. 2011). In our case, ship traffic in Calaca may come from the port in Batangas City or from the industries around the area. Industries typically use ships to transport their materials (Moldanová et al. 2009).

**Multiwavelength Absorption Analysis**

Graphical representation of the data collected by MABI (Figure 4) showed two prominent peaks, one from vehicular emission (17 Feb 2019) and another from biomass burning (30 Jan 2019). It also shows a constant source of BC that comes from biomass burning, which is represented by the red time series plot (Figure 4c). It can be said that the area around the sampling site has a constant activity of biomass burning possibly by burning dried leaves. Burning dried leaves is a common practice in rural areas in the Philippines to drive away insects such as flies and mosquitoes. As for the vehicular exhaust, BC concentrations were low during the first quarter of the sampling, December–January, while exhibiting a high-low pattern in the last half of the sampling, weekends and weekdays respectively. BC concentrations due to vehicular emission during the latter half of the sampling date were twice as high in comparison to the first half of the sampling. This is due to the fact that the last part of the December up to first quarter of January, which were included in the first half of the sampling, are Christmas holidays and the sudden increase in the last half of the sampling, which started at the last half of January, signaled the start of working and school days.

**Elemental Composition**

The results of the elemental analysis showed that sulfur was the most abundant elemental component in the PM$_{2.5}$ fraction (Figure 5). Sulfur is an element that is generally present in airborne PM because of the atmospheric conversion of secondary sulfates from SO$_2$ via oxidation and other homogeneous processes (Hopke et al. 2008). The presence of sulfur in the air can be attributed to the emissions of several significant sources such as automobile and ship vessel fuel combustion, coal-fired power plants, among other industries (Cohen 2010), which supports the presence of VBC and BBC around the area. Although the sampling site is near an ocean, Na and Cl concentrations for PM$_{2.5}$ fraction were low. This supports previous findings of similar studies indicating that Na is usually associated with large sea-salt particles suspended in the atmosphere, hence its deposition in the coarse fraction (Alastuey et al. 2016).

**Reconstructed Mass**

One of the existing methods for source type apportionment is the determination of reconstructed mass as described by Malm et al. (1994). In this method, major fine aerosol types – known as pseudo-elements (e.g., soil, salt, and sulfate) – are estimated from the obtained elemental concentrations from EDXRF analysis. Other pseudo-elements described by Malm et al. (i.e., smoke and organic carbon) were omitted due to lack of data needed for estimation. These components, along with the BC concentrations, comprise the reconstructed mass (RCM) of PM$_{2.5}$ samples as defined in Equation 2.

\[
\text{RCM} = [\text{salt}] + [(\text{NH}_4)_2\text{SO}_4] + [\text{soil}] + [\text{vehicular BC}] + [\text{biomass BC}]
\] (2)

As presented in the elemental analysis results, sulfur is the dominant inorganic species present in the PM$_{2.5}$ fraction. Elemental sulfur is primarily present in the aerosol in sulfate form, usually H$_2$SO$_4$, as a result of the oxidation of SO$_2$ gas commonly emitted from the combustion of coal, diesel, and other fossil fuels (Perrera 2018). Ammonia in the atmosphere typically neutralizes sulfuric acid, forming (NH$_4$)$_2$SO$_4$. In our calculation, it is assumed that the sulfur species in the atmosphere is completely neutralized into (NH$_4$)$_2$SO$_4$. As a result, (NH$_4$)$_2$SO$_4$ dominated the total mass of PM$_{2.5}$ fraction, contributing 61.15%, (NH$_4$)$_2$SO$_4$ in the PM$_{2.5}$ fraction. This, along with VBC and BBC, was expected because the sampling site is located near the national highway and in a residential area where burning of leaves, twigs, and other biomass is being practiced. Vehicular and biomass emissions contributed 6.28% and 6.33% of the total mass of the PM$_{2.5}$ fraction, respectively (Figure 6). CPF analysis of the sulfate component of PM$_{2.5}$ shows that sulfate primarily comes from 30–60° and 180° direction, wherein the national highway and the sea are located, respectively (Figure A1). These results support our interpretation that fuel combustion from vehicular emissions from the national highway and ship vessels from the nearby sea influences the presence of aerosol sulfate (Appendix VI).

Crustal elements (i.e., Al, Si, Ca, Ti, Fe) were also present in the PM$_{2.5}$ fraction, and their concentrations can be used to determine the estimated soil mass concentration. The soil mass concentration is estimated through the summation of these terrestrial elements and their major oxides that account for more than 85% of the total soil composition (Hopke et al. 2008). The soil component from
the calculation of the reconstructed mass accounted for 11.31% of the total PM$_{2.5}$ mass (Figure 5). This finding is also expected since the area around the sampler is not cemented, and soil dust from the ground can easily be suspended into the atmosphere through natural (e.g., wind) and anthropogenic means (e.g., physical force from sweeping, vehicular movement, walking).

CONCLUSION

In conclusion, this study provides baseline information on PM$_{10}$, PM$_{2.5}$, VBC, BBC, and elemental concentrations in the growing industrial city of Calaca, Batangas. A total of 52 filter samples, 26 for each particulate fraction (i.e., PM$_{10,2.5}$ and PM$_{2.5}$), were collected for four months, from December 2018 to March 2019.

Throughout the sampling period, the measured average mass concentrations of PM$_{10}$ and PM$_{2.5}$ were compliant to local government standards and are thus considered safe. Our analysis also showed no significant difference between weekday and weekend PM concentrations. Elemental analysis of the PM$_{2.5}$ fraction samples by EDXRF measured 14 elements, and sulfur was found to be the most dominant. Integrating results of elemental and multiwavelength BC analysis, the following sources and their respective contributions to the total PM$_{2.5}$ mass were determined: salt (0.54%), soil (11.31%), (NH$_4$)$_2$SO$_4$ (61.15%), VBC (6.33%), and BBC (6.23%). CPF determined that PM most likely comes from the 30–60° and 180° directions from the sampling site, which are possibly vehicular activity from the national highway and ship activity from the sea, respectively. Since one limitation of the study is the short sampling period that only corresponds to the amihan season of the country due to limited resources and availability of the sampler used, a longer (e.g., year-round) sampling period and more sampling sites will be helpful in determining seasonal and geographical variability of PM concentrations in the area, as well as in the implementation of a more accurate source fingerprint apportionment method.

Even though the measured PM levels are still considered safe, air sampling and monitoring of the area is still recommended in order to continuously assess the effects of the industries to the air quality in the area. The data obtained will help the local government units and policy-making bodies create laws and regulations that will help the municipality prevent the increase in air pollution. Furthermore, the results of the study will be helpful in providing awareness that pollution is not something to be taken lightly, and individual actions are needed in order to prevent casualties that air pollution might bring, as Calaca continues its rise to industrial and economic development.

ACKNOWLEDGMENTS

The authors would like to thank the National Research Council of the Philippines for providing the Gent samplers and air filters used. The authors would also like to thank the International Atomic Energy Agency for providing the MAB used in this study. The Panalytical E5 EDXRF system was provided by DOST-PNRI under the DOST-GIA Project. The authors would also like to thank Ms. Gloria Jimenez, Mr. Romeo C. Riosa, and Engr. Goldsthen Villaverde for the maintenance of the air samplers and particulate samples.

NOTES ON APPENDICES

The complete appendices section of the study is accessible at http://philjournsci.dost.gov.ph.

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APPENDIX I

Particulate Matter Concentrations

Table A1. PM\textsubscript{10} concentration for each sampling date of the week.

<table>
<thead>
<tr>
<th>Week</th>
<th>Sampling date</th>
<th>PM\textsubscript{10} load (ug/m\textsuperscript{3})</th>
<th>unc</th>
<th>PM\textsubscript{10}</th>
<th>Sampling date</th>
<th>PM\textsubscript{10} load (ug/m\textsuperscript{3})</th>
<th>unc</th>
</tr>
</thead>
<tbody>
<tr>
<td>Week 1</td>
<td>19-Dec-18</td>
<td>50.8</td>
<td>0.5</td>
<td>23-Dec-18</td>
<td>15.6</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>Week 2</td>
<td>26-Dec-18</td>
<td>39.6</td>
<td>0.8</td>
<td>30-Dec-18</td>
<td>4.6</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>Week 3</td>
<td>2-Jan-19</td>
<td>33.8</td>
<td>0.5</td>
<td>6-Jan-19</td>
<td>23.3</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>Week 4</td>
<td>9-Jan-19</td>
<td>20.5</td>
<td>0.2</td>
<td>13-Jan-19</td>
<td>28.8</td>
<td>0.4</td>
<td></td>
</tr>
<tr>
<td>Week 5</td>
<td>16-Jan-19</td>
<td>38.2</td>
<td>0.5</td>
<td>20-Jan-19</td>
<td>29.8</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>Week 6</td>
<td>23-Jan-19</td>
<td>21.2</td>
<td>0.4</td>
<td>27-Jan-19</td>
<td>40.3</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>Week 7</td>
<td>30-Jan-19</td>
<td>43.1</td>
<td>0.5</td>
<td>3-Feb-19</td>
<td>24.1</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>Week 8</td>
<td>6-Feb-19</td>
<td>30.5</td>
<td>0.5</td>
<td>17-Feb-19</td>
<td>45.0</td>
<td>0.6</td>
<td></td>
</tr>
<tr>
<td>Week 9</td>
<td>20-Feb-19</td>
<td>30.4</td>
<td>0.6</td>
<td>24-Feb-19</td>
<td>36.7</td>
<td>0.6</td>
<td></td>
</tr>
<tr>
<td>Week 10</td>
<td>27-Feb-19</td>
<td>29.5</td>
<td>0.6</td>
<td>3-Mar-19</td>
<td>46.5</td>
<td>0.6</td>
<td></td>
</tr>
<tr>
<td>Week 11</td>
<td>6-Mar-19</td>
<td>41.2</td>
<td>0.5</td>
<td>10-Mar-19</td>
<td>61.9</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>Week 12</td>
<td>13-Mar-19</td>
<td>33.4</td>
<td>0.3</td>
<td>17-Mar-19</td>
<td>38.9</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>Week 13</td>
<td>20-Mar-19</td>
<td>44.3</td>
<td>0.4</td>
<td>24-Mar-19</td>
<td>26.8</td>
<td>0.6</td>
<td></td>
</tr>
</tbody>
</table>

Table A2. PM\textsubscript{2.5} concentration for each sampling date of the week.

<table>
<thead>
<tr>
<th>Week</th>
<th>Sampling date</th>
<th>PM\textsubscript{2.5} load (ug/m\textsuperscript{3})</th>
<th>unc</th>
<th>PM\textsubscript{2.5}</th>
<th>Sampling date</th>
<th>PM\textsubscript{2.5} load (ug/m\textsuperscript{3})</th>
<th>unc</th>
</tr>
</thead>
<tbody>
<tr>
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<td>15.9</td>
<td>0.4</td>
<td>23-Dec-18</td>
<td>5.8</td>
<td>0.7</td>
<td></td>
</tr>
<tr>
<td>Week 2</td>
<td>26-Dec-18</td>
<td>11.9</td>
<td>0.3</td>
<td>30-Dec-18</td>
<td>2.6</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
<td>Week 3</td>
<td>2-Jan-19</td>
<td>11.4</td>
<td>0.5</td>
<td>6-Jan-19</td>
<td>10.2</td>
<td>0.4</td>
<td></td>
</tr>
<tr>
<td>Week 4</td>
<td>9-Jan-19</td>
<td>5.0</td>
<td>0.4</td>
<td>13-Jan-19</td>
<td>7.5</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
<td>Week 5</td>
<td>16-Jan-19</td>
<td>10.8</td>
<td>0.3</td>
<td>20-Jan-19</td>
<td>10.9</td>
<td>0.4</td>
<td></td>
</tr>
<tr>
<td>Week 6</td>
<td>23-Jan-19</td>
<td>5.8</td>
<td>0.6</td>
<td>27-Jan-19</td>
<td>8.3</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>Week 7</td>
<td>30-Jan-19</td>
<td>22.3</td>
<td>0.1</td>
<td>3-Feb-19</td>
<td>3.3</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>Week 8</td>
<td>6-Feb-19</td>
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<td>17-Feb-19</td>
<td>15.1</td>
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<tr>
<td>Week 9</td>
<td>20-Feb-19</td>
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<td>0.9</td>
<td>24-Feb-19</td>
<td>13.7</td>
<td>0.4</td>
<td></td>
</tr>
<tr>
<td>Week 10</td>
<td>27-Feb-19</td>
<td>9.7</td>
<td>0.3</td>
<td>3-Mar-19</td>
<td>9.4</td>
<td>0.5</td>
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</tr>
<tr>
<td>Week 11</td>
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<td>0.7</td>
<td>10-Mar-19</td>
<td>11.8</td>
<td>0.7</td>
<td></td>
</tr>
<tr>
<td>Week 12</td>
<td>13-Mar-19</td>
<td>11.3</td>
<td>0.8</td>
<td>17-Mar-19</td>
<td>21.3</td>
<td>0.2</td>
<td></td>
</tr>
<tr>
<td>Week 13</td>
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<td>0.4</td>
<td>24-Mar-19</td>
<td>11.7</td>
<td>0.3</td>
<td></td>
</tr>
</tbody>
</table>

APPENDIX II

Weekday and Weekend Comparison

Table A3. T-test for two samples with unequal variances for both PM\textsubscript{10} and PM\textsubscript{2.5}.

<table>
<thead>
<tr>
<th>Month</th>
<th>PM\textsubscript{10}</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>Critical</td>
<td>P-value</td>
</tr>
<tr>
<td>December</td>
<td>4.30</td>
<td>0.04</td>
</tr>
<tr>
<td>January</td>
<td>2.57</td>
<td>0.83</td>
</tr>
<tr>
<td>February</td>
<td>3.18</td>
<td>0.49</td>
</tr>
<tr>
<td>March</td>
<td>2.78</td>
<td>0.91</td>
</tr>
</tbody>
</table>

\(^1\text{T-test was done at a 95% confidence interval.}\)
APPENDIX III

Elemental and Chemical Concentrations

Table A4. Summary statistics of elements and chemicals detected in the fine fraction.

<table>
<thead>
<tr>
<th>Element</th>
<th>n</th>
<th>Mean conc. (ng/m$^3$)</th>
<th>Min. (ng/m$^3$)*</th>
<th>Max. (ng/m$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na</td>
<td>26</td>
<td>22.59</td>
<td>108.36</td>
<td>189.02</td>
</tr>
<tr>
<td>Al</td>
<td>26</td>
<td>58.03</td>
<td>7.67</td>
<td>182.68</td>
</tr>
<tr>
<td>Si</td>
<td>26</td>
<td>256.04</td>
<td>51.25</td>
<td>691.68</td>
</tr>
<tr>
<td>S</td>
<td>26</td>
<td>1573.63</td>
<td>759.31</td>
<td>3074.91</td>
</tr>
<tr>
<td>Cl</td>
<td>26</td>
<td>41.19</td>
<td>71.72</td>
<td>298.30</td>
</tr>
<tr>
<td>K</td>
<td>26</td>
<td>138.16</td>
<td>15.02</td>
<td>480.44</td>
</tr>
<tr>
<td>Ca</td>
<td>26</td>
<td>73.30</td>
<td>18.58</td>
<td>233.34</td>
</tr>
<tr>
<td>Se</td>
<td>26</td>
<td>0.86</td>
<td>5.62</td>
<td>10.32</td>
</tr>
<tr>
<td>Ti</td>
<td>26</td>
<td>15.28</td>
<td>8.85</td>
<td>38.50</td>
</tr>
<tr>
<td>Mn</td>
<td>26</td>
<td>37.81</td>
<td>11.43</td>
<td>83.81</td>
</tr>
<tr>
<td>Fe</td>
<td>26</td>
<td>119.04</td>
<td>0.95</td>
<td>319.24</td>
</tr>
<tr>
<td>Ni</td>
<td>26</td>
<td>5.32</td>
<td>9.93</td>
<td>15.40</td>
</tr>
<tr>
<td>Cu</td>
<td>26</td>
<td>0.68</td>
<td>0.86</td>
<td>3.76</td>
</tr>
<tr>
<td>Zn</td>
<td>26</td>
<td>12.53</td>
<td>0.89</td>
<td>52.93</td>
</tr>
<tr>
<td>PM</td>
<td>26</td>
<td>10615.69</td>
<td>2632.11</td>
<td>22273.13</td>
</tr>
<tr>
<td>VBC</td>
<td>26</td>
<td>671.46</td>
<td>88.11</td>
<td>2082.80</td>
</tr>
<tr>
<td>BBC</td>
<td>26</td>
<td>666.39</td>
<td>348.68</td>
<td>1804.00</td>
</tr>
</tbody>
</table>

*Non-zero concentrations

APPENDIX IV

Pseudo-elemental Concentrations

Table A5. Pseudo-elemental concentrations in the PM$_{2.5}$ fraction.

<table>
<thead>
<tr>
<th>Sampling date</th>
<th>PM$_{2.5}$ (µg/m$^3$)</th>
<th>soil (µg/m$^3$)</th>
<th>(NH$_4$)$_2$SO$_4$ (µg/m$^3$)</th>
<th>Biomass BC (µg/m$^3$)</th>
<th>Vehicular BC (µg/m$^3$)</th>
<th>RCM (µg/m$^3$)</th>
<th>RCM %</th>
</tr>
</thead>
<tbody>
<tr>
<td>12/19/2018</td>
<td>15.94</td>
<td>1.20</td>
<td>7.69</td>
<td>0.70</td>
<td>0.21</td>
<td>9.80</td>
<td>61.50</td>
</tr>
<tr>
<td>12/23/2018</td>
<td>5.79</td>
<td>0.20</td>
<td>3.76</td>
<td>0.35</td>
<td>0.00</td>
<td>4.31</td>
<td>74.37</td>
</tr>
<tr>
<td>12/26/2018</td>
<td>11.90</td>
<td>0.35</td>
<td>5.38</td>
<td>0.74</td>
<td>0.54</td>
<td>7.01</td>
<td>58.90</td>
</tr>
<tr>
<td>12/30/2018</td>
<td>2.63</td>
<td>0.00</td>
<td>3.13</td>
<td>0.51</td>
<td>0.00</td>
<td>3.65</td>
<td>138.53</td>
</tr>
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<td>01/02/2019</td>
<td>11.43</td>
<td>0.24</td>
<td>3.61</td>
<td>0.66</td>
<td>0.00</td>
<td>4.51</td>
<td>39.43</td>
</tr>
<tr>
<td>01/06/2019</td>
<td>10.24</td>
<td>0.42</td>
<td>7.13</td>
<td>0.55</td>
<td>0.79</td>
<td>8.90</td>
<td>86.90</td>
</tr>
<tr>
<td>01/09/2019</td>
<td>5.02</td>
<td>0.25</td>
<td>3.53</td>
<td>0.43</td>
<td>0.00</td>
<td>4.21</td>
<td>83.88</td>
</tr>
<tr>
<td>01/13/2019</td>
<td>7.45</td>
<td>0.36</td>
<td>4.59</td>
<td>0.38</td>
<td>0.09</td>
<td>5.42</td>
<td>72.74</td>
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<tr>
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<td>0.91</td>
<td>7.84</td>
<td>0.91</td>
<td>1.35</td>
<td>11.02</td>
<td>101.73</td>
</tr>
<tr>
<td>02/20/2019</td>
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<td>5.44</td>
<td>0.55</td>
<td>1.23</td>
<td>8.06</td>
<td>73.79</td>
</tr>
<tr>
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<td>0.60</td>
<td>0.00</td>
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</tr>
<tr>
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<td>1.19</td>
<td>7.95</td>
<td>0.36</td>
<td>0.74</td>
<td>10.23</td>
<td>123.85</td>
</tr>
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<td>5.89</td>
<td>1.80</td>
<td>1.20</td>
<td>10.18</td>
<td>45.70</td>
</tr>
<tr>
<td>02/03/2019</td>
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<td>0.29</td>
<td>4.13</td>
<td>0.42</td>
<td>0.83</td>
<td>5.67</td>
<td>173.19</td>
</tr>
</tbody>
</table>
APPENDIX V

Meteorological Data

Table A6. Wind direction and speed values throughout the sampling period.

<table>
<thead>
<tr>
<th>Date</th>
<th>Time</th>
<th>Wind speed (m/s)</th>
<th>Wind direction</th>
<th>Wind direction (°)</th>
</tr>
</thead>
<tbody>
<tr>
<td>02/06/2019</td>
<td>00:00 - 06:00</td>
<td>0.56</td>
<td>NE</td>
<td>45</td>
</tr>
<tr>
<td>02/06/2019</td>
<td>06:00 - 12:00</td>
<td>1.94</td>
<td>NNE</td>
<td>22.5</td>
</tr>
<tr>
<td>02/06/2019</td>
<td>12:00 - 18:00</td>
<td>2.22</td>
<td>NE</td>
<td>45</td>
</tr>
<tr>
<td>02/06/2019</td>
<td>18:00 - 00:00</td>
<td>1.39</td>
<td>NE</td>
<td>45</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>00:00 - 06:00</td>
<td>1.39</td>
<td>NE</td>
<td>45</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>06:00 - 12:00</td>
<td>1.67</td>
<td>NE</td>
<td>45</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>12:00 - 18:00</td>
<td>1.39</td>
<td>NE</td>
<td>45</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>18:00 - 00:00</td>
<td>0.83</td>
<td>NE</td>
<td>45</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>00:00 - 06:00</td>
<td>0.00</td>
<td>N</td>
<td>0</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>06:00 - 12:00</td>
<td>0.83</td>
<td>NNE</td>
<td>22.5</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>12:00 - 18:00</td>
<td>0.83</td>
<td>NNE</td>
<td>22.5</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>18:00 - 00:00</td>
<td>0.83</td>
<td>N</td>
<td>0</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>00:00 - 06:00</td>
<td>1.39</td>
<td>NE</td>
<td>45</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>06:00 - 12:00</td>
<td>1.39</td>
<td>NE</td>
<td>45</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>12:00 - 18:00</td>
<td>1.39</td>
<td>NE</td>
<td>45</td>
</tr>
<tr>
<td>02/17/2019</td>
<td>18:00 - 00:00</td>
<td>1.39</td>
<td>NE</td>
<td>45</td>
</tr>
<tr>
<td>02/20/2019</td>
<td>00:00 - 06:00</td>
<td>0.00</td>
<td>N</td>
<td>0</td>
</tr>
<tr>
<td>02/20/2019</td>
<td>06:00 - 12:00</td>
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APPENDIX VI

Conditional Probability Values

Table A7. CPF values for PM_{10}.

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Table A8. CPF values for PM_{2.5}.

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Table A9. CPF values for sulfate source in the PM$_{2.5}$ fraction.

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Figure A1. Upper 50% conditional probability function (CPF) graphs for the sulfate component of PM$_{2.5}$ obtained from the wind flow behavior of Calaca, Batangas.